Very fast YbxY1-xAl03 single-crystal scintillators

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Very fast $Yb_xY_{1-x}AlO_3$ single-crystal scintillators

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Scintillation, radioluminescence, photoluminescence, and thermoluminescence characteristics of Czochralski-grown $Yb_xY_{1-x}AlO_3$ single crystals for $x=0.02-0.45$ are described. Under x-ray excitation, the intensity of Yb^{3+} charge-transfer luminescence is increasing with x and reaches at room temperature more than 11% of $Bi_4Ge_3O_{12}$ (BGO) for $x=0.45$, while at 80 K it is over 170% of BGO for $x\geqslant 0.3$. At room temperature, scintillation decay shows a dominant decay time of 0.8–0.9 ns for $x\geqslant 0.3$ and no significant slow components were detected. The photoluminescence decay time is tuned by temperature and Yb concentration from tens of nanoseconds at 80 K down to about 0.8 ns at 295 K. Thermoluminescence glow curve below room temperature evidences very low concentration of shallow traps compared to the Ce-doped $YAlO_3$ scintillator. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1645987]

Luminescence and scintillation of Yb-doped materials became of interest in recent years[1–3] due to their possible application for neutrino detection in high-energy physics experiments.[4] After just two studies in this field in the 1970s,[5,6] selected features of the charge-transfer (CT) luminescence of Yb^{3+} in various matrices were reported.[1] More systematic attention was paid to Yb-doped $Y_3Al_5O_{12}$ (YAG)[2,3] due to the well-established technology of the crystal growth and simultaneous study of the Yb^{3+} infrared $4f$-$4f$ luminescence.[7] Yb-doped $YAlO_3$ (YAP) became noticed as well: a light yield of Yb8%:YAP was reported[2] of 7.5% of BGO and luminescence characteristics under vuv excitation were reported very recently for the same sample as well.[8] Single-crystal rods of the Yb-rich $Yb_xY_{1-x}AP$ (up to $x=0.2$) were prepared by the micropulling-down method[9] and luminescence characteristics measured as well, showing in accordance with Ref. 8 the edge of the CT luminescence excitation around 250 nm and the double peak emission spectrum at about 350+520 nm. Growth and luminescence properties of high-quality Czochralski-grown single crystals of $Yb_xY_{1-x}AlO_3$ single crystals for $x=0.02-0.45$ (65-mm-long crystals of 20 mm diam) were reported very recently[10] showing an interesting anomalous behavior for the temperature dependence of CT luminescence and its dependence on Yb concentration. Despite the partial concentration quenching effect evidenced for $x\geqslant 0.3$ from the luminescence decay, the temperature quenching appeared less intense with increasing Yb content and a hypothesis was given about restricted relaxation of the CT excited state due to decreasing lattice constant in Yb-rich YAP crystal.

The aim of this letter is to report the scintillation characteristics of Czochralski-grown single crystals of $Yb_xY_{1-x}AlO_3$ single crystals for $x=0.02-0.45$ and compare their parameters with those of BGO and BaF_2 scintillators.

Photoluminescence (PL) and radioluminescence (RL) spectra and decays were measured with a modified Spectrofluorometer 199S, Edinburgh Instrument, using excitation with a steady state or ns pulsed hydrogen discharge flashlamps, x-ray tube, and ^{22}Na radioisotope, respectively (for other details, see Refs. 9 and 10). Emission spectra were corrected for the experimental distortions and true decay times were extracted from the decay curves using a deconvolution procedure (SpectraSolve software of LASTEK Ltd.). Wavelength-resolved thermally stimulated luminescence (TSL) measurements were performed in the 10–310 K range after x-ray irradiation at 10 K (by a Philips 2274 x-ray tube operated at 20 kV), with a homemade apparatus allowing the detection of the TSL signal both as a function of temperature and wavelength. The signal detection was performed by a monochromator coupled to a CCD (Jobin Yvon Spectrum 3000) operating in the 280–710-nm interval. Details of the crystal growth by the Czochralski method can be found in Ref. 10. $Yb_xY_{1-x}AlO_3$ crystals with $x=0.02$, 0.1, 0.2, 0.3, and 0.45 were studied; hereafter, the samples will be labeled as Yb-x.

RL spectra are shown for the limit Yb concentrations in Fig. 1 at room temperature (RT) and compared in the absolute scale with the standard BGO sample of the same dimensions. Identically to PL,[8–10] RL spectra of Yb-0.02 and Yb-

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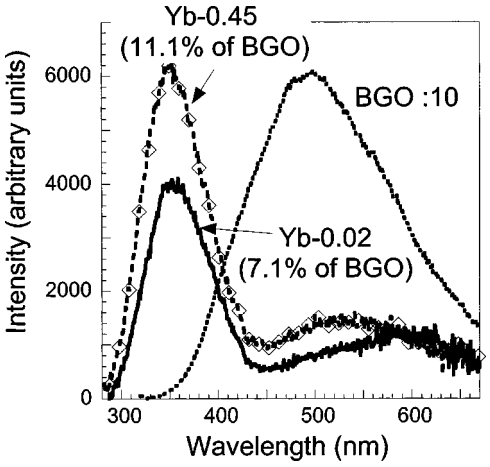

FIG. 1. Radioluminescence of Yb:YAP and BGO at RT. Excitation by x-ray tube, 35 kV, 15 mA. Quantitative comparison with respect to BGO is provided by the calculation of spectra integrals.

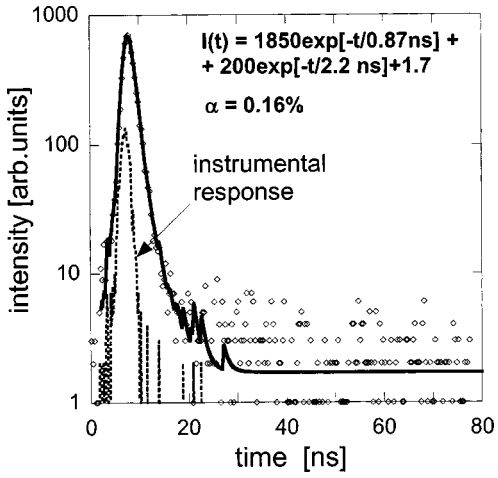

FIG. 3. Scintillation decay of Yb-0.45 at room temperature. Excitation by 511 keV photons of ^{22}Na radioisotope, spectrally unresolved. The two-exponential approximation is given by a solid line: convolution of the two-exponential function in the figure with the instrumental response given by a dashed line. The coefficient alpha related to the relative amplitude of the superslow components calculated according Ref. 12 is also given.

0.45 are governed by the Yb^{3+} CT luminescence peaking at 350+530 nm. In the former sample, a parasitic luminescence is noticed in the red spectral region, which was mentioned in the YAP host earlier.[11] Interestingly, total RL intensity (integrated emission spectra) is monotonously increasing with the Yb concentration and reaches more than 11% of the BGO intensity for Yb-0.45. The temperature dependence of the RL intensity is given in Fig. 2 and normalized to that of BGO at RT. For higher Yb concentration, the RL intensity is higher over the entire temperature interval measured (80–300 K) and exceeds 170% of BGO for $x \geqslant 0.3$ at 80 K. At low temperatures, the PL decay curves experience the well-observable concentration quenching effect[10] for $x \geqslant 0.3$, which results in a pronounced nonexponentiality and speeding-up of the decay curve in its initial part. Due to the nonexponential decay mentioned, 1/*e* decay time was evaluated over the entire temperature range for Yb-0.02 and Yb-0.3 samples; see the inset of Fig. 2. While at low temperatures the values of 1/*e* decay times differ significantly due to the mentioned concentration quenching effect, around RT they closely resemble each other (and the decay curve can be well approximated by a single exponential function for any *x*), because the thermal quenching mechanism becomes dominant. It is worth noting that with increasing Yb concentration, we obtained at the same time increasing RL intensity but decreasing 1/*e* PL decay time.

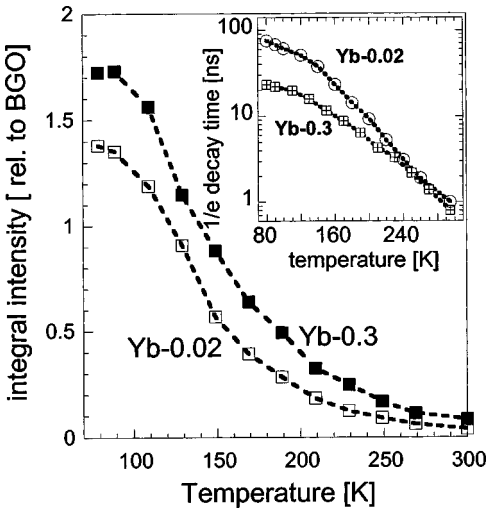

FIG. 2. Temperature dependence of the radioluminescence intensity (calculated as RL spectra integrals at each temperature). Excitation by x-ray tube, 35 kV, 15 mA. *y* axis is calibrated in units of BGO RT radioluminescence intensity obtained from quantitative comparison at RT—see Fig. 1. In the inset, the temperature dependence of photoluminescence 1/*e* decay times is given for Yb-0.02 and Yb-0.3 samples, with an excitation of 230 nm.

Scintillation decay of the Yb-0.45 sample is given in Fig. 3. The two-exponential approximation shows the dominant decay component of 0.87 ns decay time followed by the low-in-amplitude component of about 2.2 ns decay time. Evaluation of the eventual very slow decay components responsible for the background increase due to repetitive excitation by the ^{22}Na isotope (about 20 kHz), the so-called coefficient alpha introduced in the description of $PbWO_4$ characteristics,[12] provides an extremely low value of about 0.16%, which confirms the practical absence of any such slower processes in the scintillation decay. At RT, scintillation decay of all the samples closely resembles the PL one.

To monitor shallow trapping states, which can effectively delay radiative recombination of migrating electrons and holes due to retrapping processes, wavelength-resolved TSL measurements were performed on Yb-0.3 after irradiation at 10 K (Fig. 4, curve *a*). Two glow peaks at 33 and 55 K and a very weak one at around 150 K were observed. At temperatures higher than 70 K, the TSL intensity is a few orders weaker with respect to that of the sample previously studied by us, which was YAP:1%Ce, 1000 ppm Zr,[13] featuring the optimized scintillation properties (here also reported as curve *b* of Fig. 4). Considering the irradiation dose mentioned in the caption of Fig. 4, the integral of the TSL glow curves in Fig. 4 points to at least a one order lower concentration of (TSL-active) shallow traps in Yb-0.3 with respect to the optimized YAP:Ce,Zr. This result is coherent with the observed fast scintillation decay described above and the very low alpha coefficient calculated for Yb-0.3: in Ce-doped YAP, it is usually about 2% or higher.[13] The close similarity of the PL and TSL spectra evidences the crucial role of CT-Yb^{3+} centers in both the prompt and delayed radiative

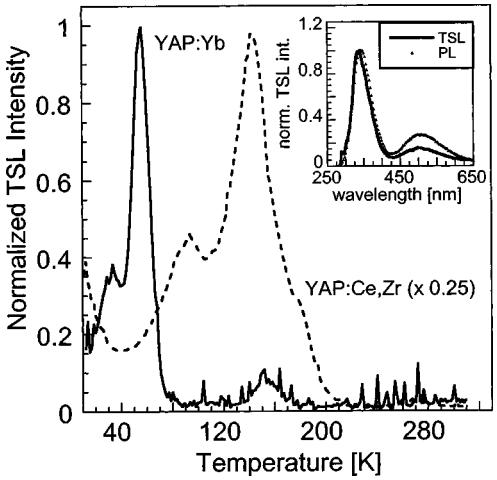

FIG. 4. TSL glow curve after x-ray irradiation of Yb-0.3 sample (curve *a*) and of YAP:1%Ce, 1000 ppm Zr (curve *b*) obtained by integration of the wavelength resolved measurements over the 280–710-nm emission spectral range. The curves are normalized to the maximum TSL intensity of Yb-0.3. Heating rate=0.1 K/s. The x-ray dose imparted to Yb-0.3 is approximately *14 times higher* with respect to that imparted to YAP:Ce,Zr. In the inset, for Yb-0.3 the TSL emission spectrum in the 50–60-K range and PL emission spectrum under excitation at 230 nm at 50 K are reported.

recombination processes in $Yb_xY_{1-x}AlO_3$ crystals.

We conclude that the obtained scintillation characteristics of Yb-rich ($x \geqslant 0.3$) $Yb_xY_{1-x}AlO_3$ are very favorable: since all the radioluminescence intensity [11.1% of BGO for $x=0.45$; BGO light yield (LY) is reported in the literature between 8000 and 9000 phot/MeV (Ref. 14)] is released with a decay time of about 0.8–0.9 ns as indicated by the scintillation decay, the LY of this fast component is expected to be about 70% of the fast BaF_2 scintillation component (reported LY of about 1400–1500 phot/MeV, decay time of 0.6–0.8 ns related to the cross-luminescence component peaking between 180 and 220 nm).[14] However, contrary to BaF_2, in $Yb_xY_{1-x}AlO_3$ no slower decay processes are present and the emission spectrum is favorably placed within the near-uv visible spectral range. Furthermore, $Yb_{0.45}Y_{0.55}AlO_3$ is of much higher density (6.64 g/cm^3), and Yb:YAP is of higher chemical and mechanical stability with respect to BaF_2. It is also worth noting that among known and practically important scintillation materials, there is none which would enable temperature tuning of its scintillation response within the range of sub-ns–several tens of ns decay times, which is in principle offered by this system within 80–300 K as demonstrated by the temperature dependence of 1/*e* PL decay times. Possible comparison could be made only with the heavily trivalent ion-doped $PbWO_4$, which also shows at RT subnanosecond scintillation decay free of longer components.[15] However, its RL intensity at room temperature is less than 1% of BGO, i.e., at least one order of magnitude lower with respect to the present $Yb_{0.45}Y_{0.55}AlO_3$ crystal.

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